LAB-SCALE STUDIES OF OXYFUEL COMBUSTION

A. Roisman, A. Suriyawong, M. Holtmeyer, S.A. Skeen, P. Biswas and R.L. Axelbaum

Energy, Environment and Chemical Engineering Department
Washington University in Saint Louis, Saint Louis, Missouri 63130
Telephone and Fax number of PI (RLA): 314-935-7560, 314- 935-4014 (fax)

Grant Number: DE-FG26-05NT4253
Performance Period: 8/1/05-2/28/07

OBJECTIVES

As part of the Global Climate Change Initiative and the need to manage green house gases, there is strong interest in developing a portfolio of technologies for minimizing emissions of carbon dioxide from coal-fired systems. The low concentration of carbon dioxide in the flue gas of conventional coal-fired plants makes it difficult to develop cost effective means of capturing and sequestering carbon dioxide. An operational scheme based on flue-gas recycling and pure oxygen can result in greater than 95% carbon dioxide in the coal combustor exhaust. This will improve carbon dioxide recovery and also potentially minimize NOx, SOx, and trace species emissions. Past studies have focused on replacing the nitrogen in the air with CO_2 from the exhaust gas. While this approach is effective at increasing the CO_2 concentration in the flue gas, the potential for even greater improvements in the overall cycle is possible. This can be appreciated by recognizing that since there are actually three streams (oxygen, CO_2, and coal), there are an unlimited number of ways these three streams can be mixed, and the resulting products can vary dramatically.

Thus motivated, the goal of this program is determining how to best implement oxycoal combustion with flue gas recirculation and sorbent processes to minimize emissions and maximize combustion efficiency and the concentration of CO_2 in the flue gas. The optimum approach to mixing of the coal, oxygen and CO_2 and sorbents is being evaluated and the effect of this mixing and the enriched carbon dioxide environment on emission of NOx, SOx, PM 2.5 and mercury is being studied as well.

Experiments are being conducted in a drop tube furnace to understand the role of CO_2 without considering mixing. In addition, a 40 kW laboratory-scale oxycoal-fired burner has been constructed and is being used to test mixing concepts for a wide range of feed conditions and temperatures. Some sorbent studies are also being conducted at the NETL pilot scale facility.

ACCOMPLISHMENTS TO DATE

Primary conclusions of results to date are summarized below. Detailed information can be obtained in the references cited.

1) Replacing nitrogen with carbon dioxide can lead to a smaller size distribution of submicron particles and lower number concentration. The large specific heat of carbon dioxide lowers flame temperature and this slows the vaporization rate, leading to a delay in the onset of nucleation and less particles. This also implies less time for coagulation and condensation, which results in a smaller size. When the oxygen concentration was increased to yield a similar flame temperature to that of air-fired combustion, the size distribution was similar to that for air-fired combustion.

2) A study of mercury emission was conducted for 20% O_2/80% CO_2 and 25% O_2/75% CO_2. The results revealed that the total gaseous mercury concentration was similar to that of air-fired

combustion, as was the ratio of oxidized to elementary mercury. The results suggest that carbon dioxide does not have a strong impact on mercury speciation.

3) Mercury was found to exist only in the elemental form when the carrier gas is either air or a mixture of O_2-CO_2. However, in the presence of chlorine species, oxidized mercury was found in higher concentrations in the O_2-CO_2 system than in the conventional (air) system. Water vapor did not affect mercury speciation in either O_2-CO_2 or conventional systems. However, it did play an important role on oxidation of mercury when chlorine species were present.

4) Submicrometer and ultrafine particles produced by coal combustion were found to have a bipolar charge distribution at the outlet of the combustor, which is slightly skewed towards positive charge. This appears to be relatively independent of combustion gas composition. In addition, electrostatic precipitators required higher energy inputs for O_2-CO_2 systems to produce the same number concentration of ions as O_2-N_2 systems. Furthermore, the penetration of submicrometer and ultrafine particles was an order of magnitude higher in O_2-CO_2 systems as compared to O_2-N_2 systems. These findings should be taken into consideration during design of electrostatic precipitator systems used with O_2-CO_2 coal combustion.

5) A model of soot inception limits under oxy-fuel combustion conditions was developed and validated with gaseous fuels. These results indicate that the primary parameters controlling soot inception are local temperature, local C/O ratio and residence time.

6) Flame stability in a Type I laboratory scale pulverized coal combustor was quantified as a function of inert type and oxygen concentration in both the primary oxidizer and secondary oxidizer streams. Flame stability was characterized by the primary oxidizer Reynolds number at flame blow-off. Inert exchange (removing inert from the secondary oxidizer and increasing inert in the primary) was examined as an approach to oxy-coal combustion, and its effects on flame stability were compared with the effects of conventional oxy-coal combustion. Results showed that 30% O2 by mole is required when using CO_2 as the inert to obtain stability results similar to conventional coal air combustion. Inert exchange flames were shown to have improved flame stability when compared with conventional coal air combustion even though the oxygen concentration in the primary oxidizer stream was substantially reduced. This has promising implications for NOx control and during the next period NOx measurements will be performed.

7) The performance of titanium dioxide with UV irradiation for mercury capture was evaluated in three systems: a bench-scale coal combustor, a slip stream from a pilot scale system, and a pilot scale system. The TiO_2 sorbent exhibited the highest efficiency in the bench scale system because this system allowed for the longest residence time for UV irradiation. The sorbent also demonstrated a high capture efficiency, but with a lower mercury-to-TiO_2 ratio, in the slip stream system. The flue-gas composition of the slip stream was similar to that of full-scale facilities, indicating that the performance of TiO_2 is favorable under realistic operating conditions. Tests conducted with the pilot scale system indicated that design considerations are needed to maximize the effectiveness of TiO_2 for mercury capture.

PAPERS PUBLISHED, CONFERENCE PRESENTATIONS, STUDENTS SUPPORTED

Papers and Presentations:

1. Suriyawong, A., Gamble, M., Lee, M.-H., Axelbaum, R.L. and Biswas, P., “Submicron Particle Formation and Mercury Speciation under Oxygen-Carbon Dioxide Coal Combustion,” Twenty-Second Annual International Pittsburgh Coal Conference, Pittsburgh, PA, Sept. 12-15, 2005.
2. Kumfer, B.M., Biswas, P. and Axelbaum, R.L., “Oxy-Combustion: Novel Strategies for Improving Combustion and Multi-Pollutant Control,” Twenty-Second Annual International Pittsburgh Coal Conference, Pittsburgh, PA, Sept. 12-15, 2005.
3. Suriyawong, A., Smallwood, M., Noel, J.D., Lee, M.H., Giammar, D.E., and Biswas, P. “A Strategic Approach for Optimizing the Use of Sorbents for Mercury Removal

from Coal-burning Utilities," The Air and Waste Management Association's 98th Annual Conference and Exhibition, Minneapolis, MN, 2005.

4. Suriyawong, A., Lee, M.H., and Biswas P. "Submicrometer Particle Formation and Mercury Emission under Enriched Oxygen Coal Combustion," Student Paper and Poster Competition, the Air and Waste Management Association's 98th Annual Conference and Exhibition, Minneapolis, MN, 2005.
5. Biswas, P., Suriyawong, A., Smallwood, M., Noel, J.D., Lee, M.H., Giammar, D.E., and Biswas, P. "Nanostructured-sorbents for Heavy Metals Emissions Control-A Review" American Chemical Society (ACS) 2005 Annual Conference, San Diego, CA, 2005.
6. Giammar, D.E., Noel, J.D., Smallwood, M., Suriyawong, A., Lee, M.H., and Biswas, P. "Nanostructured Sorbents for Control of Mercury Emissions from Coal Combustion," The Association of Environmental Engineering and Science Professors (AEESP), Syracuse, NY, 2005.
7. Suriyawong, A., Gamble, M.D., Lee, M., Axelbaum, R.L., and Biswas, P., "Submicrometer Particle Formation and Mercury Speciation under O_2-CO_2 Coal Combustion," *Energy & Fuels* 20 (6), 2357 -2363, 2006.
8. Skeen, S.A., Kumfer, B.M., and Axelbaum, R.L., "An Experimental and Theoretical Approach to Soot Particle Inception in Laminar Diffusion Flames," 7th International Aerosol Conference, September 10-15, 2006, St. Paul, MN.
9. Axelbaum, R.L. (Invited), "Oxy-Fuel Combustion: New opportunities for non-premixed combustion brought about by global warming," 22nd Annual Symposium of the Israeli Section of the Combustion Institute. Tel Aviv, Israel December 21, 2006.
10. Suriyawong, A., Hogan C., Jiang, J., and Biswas P., "Charge Fraction and Electrostatic Capture of Ultrafine and Submicrometer Particles Formed under O_2-CO_2 Coal Combustion," *Fuel* 87(6), 673-682, 2007.
11. Suriyawong, A., Skeen, S., Roisman, A., Axelbaum, R., and Biswas P., "Characterization of Aerosols, and Mercury Speciation under Enriched Oxygen Coal Combustion," AIChE Annual Meeting, Salt Lake City, UT, Nov. 4-9, 2007.
12. Holtmeyer, M., Roisman, A., Skeen, S.A. and Axelbaum, R.L., and Biswas P., "Flame Stability in Oxygen-Enhanced Combustion of Coal," AIChE Annual Meeting, Salt Lake City, UT, Nov. 4-9, 2007.
13. Kumfer, B.M., Skeen, S.A. and Axelbaum, R.L., "Soot Inception Limits in Laminar Diffusion Flames with Application to Oxy-Fuel Combustion," *Combustion and Flame*, in-press (2008).

Students:

Achariya Suriyawong (Ph.D.)
Scott A. Skeen (Ph.D.)
Ari Roisman (M.S.)
Melissa Holtmeyer (Ph.D.)